

## Abstract

Visual imperfections in multilayer films can be grouped into 3 categories: very small distortions, referred to as hazy film; distortions that are large enough to exhibit distinct patterns without magnification, referred to as melt fracture and interfacial instability; and large localized distortions referred to as gels. This paper will review root causes and options to minimize gauge variation of individual layers in co-extruded films. The causes and strategies apply to both cast and blown film extrusion.

## Optical Properties

The five most common terms used to describe optical properties of film are illustrated in Figure 1. Hazy film is often referred to as cloudy film. The standard test for haze is to measure the amount of light that is deflected by more than  $2.5^\circ$  when it passes through film. There are two types of haze: **Internal Haze** and **External Haze**. Internal haze is caused by scattering of light by large crystalline structures called lamellae. External haze is actually a very mild version of melt fracture or die lines. The best way to reduce internal haze is to reduce the size and frequency of crystals that scatter light. The best way to minimize external haze is to make the film surface smoother.

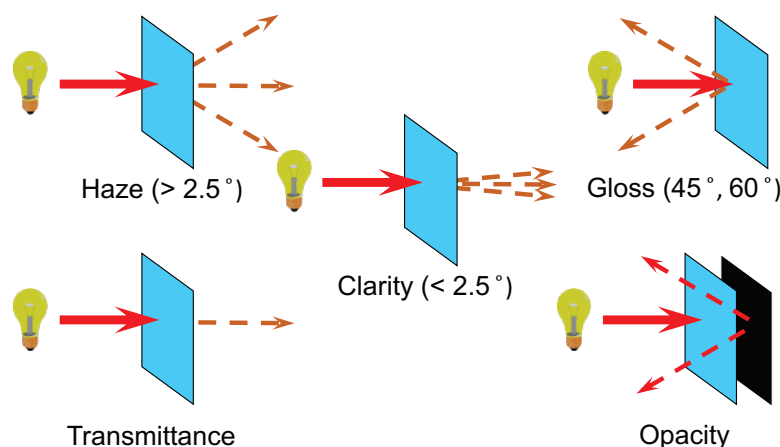


Figure 1: Optical property definitions

Film may also appear hazy due to very small distortions at the interface between adjacent layers in coextruded film. Such distortions are referred to as interfacial instability. Very small distortions are often difficult to detect without magnification. Problems may only be detected when the barrier performance of the film structure is below standard. This occurs when there are localized regions within the barrier layer that are much thinner than required to provide the required barrier performance.

## Strategies to Reduce Internal Haze

- |                       |   |
|-----------------------|---|
| Raw Material          | <ul style="list-style-type: none"> <li>• Select formulation with better optical properties:             <ul style="list-style-type: none"> <li>• Less crystalline (lower density polyolefin);</li> <li>• More amorphous (more linear molecular structure);</li> <li>• Lower average molecular rate (lower MFI / MFR);</li> <li>• Select inorganic additives with smaller particle size or better dispersion.</li> </ul> </li> </ul>                         |
| Processing Conditions | <ul style="list-style-type: none"> <li>• Increase film cooling rate (colder casting drum, colder or higher chilled air circulation rates).</li> <li>• Decrease temperature of melt:             <ul style="list-style-type: none"> <li>• Decrease die lip or die temperature;</li> <li>• Decrease melt temperature by adjusting extruder temperature profile;</li> <li>• Decrease extruder screw speed and line speed (less output).</li> </ul> </li> </ul> |
| Equipment             | <ul style="list-style-type: none"> <li>• Check and repair cooling systems that solidify melt.</li> <li>• Recalibrate blenders if required to correct formulation.</li> </ul>  |

### Strategies to Reduce External Haze (Dull, Low Gloss Surface)

- |                       |  |
|-----------------------|--|
| Raw Material          | <ul style="list-style-type: none"> <li>• Reduce shear viscosity by selecting formulation with higher MFI / MFR.</li> <li>• Reduce COF of metal surfaces by adding polymer processing aid (PPA).</li> </ul>   |
| Processing Conditions | <ul style="list-style-type: none"> <li>• Decrease film cooling rate so that film surface will be smoother before it solidifies:             <ul style="list-style-type: none"> <li>• Increase casting drum temperature (cast film);</li> <li>• Increase air temperature (blown film);</li> <li>• Increase air gap (cast film);</li> <li>• Decrease cooling air velocity (blown film);</li> <li>• Increase melt temperature by adjusting extruder temperature profile;</li> <li>• Increase extruder screw speed and line speed (more output);</li> <li>• Increase die temperature.</li> </ul> </li> </ul> |
| Equipment             | <ul style="list-style-type: none"> <li>• Clean downstream surfaces that can scuff film surface.</li> </ul>   |

### Good Layer Convergence Criteria

The coextrusion process relies on adjacent materials having similar flow characteristics so that they form a stable interface while flowing through the extrusion die. The closer the adjacent material viscosities match at production shear rates, the easier it will be to merge the flow of the materials from two flow channels into one larger flow channel. The velocity profile in the individual channels will be a symmetrical laminar flow pattern. The velocity profile in the combined flow channel will be laminar, but almost never symmetrical. Shear stress will be at a minimum in the center of the flow channels. The melted polymer that forms the thinner portion of the combined structure is referred to as the Minor Layer. The melted polymer that forms the thicker portion of the combined structure is referred to as the Major Layer. Refer to Figure 2 for an illustration of two layers that merge together with good layer convergence inside a cast or blown film die.

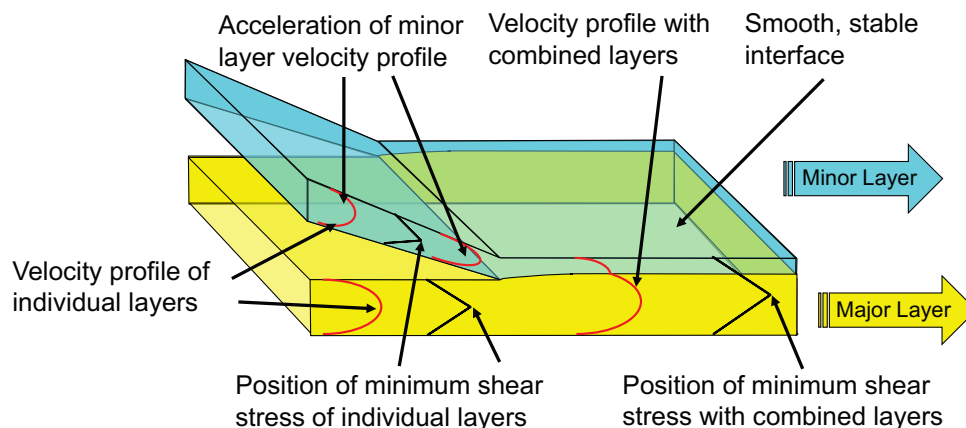


Figure 2: Melt flow patterns with good layer convergence in coextrusion

### Very Long Wave Interfacial Instability

The problem is common with cast film die blocks that assemble multilayer structures prior to entering single manifold cast film dies. The magnitude of the problem is affected by the difference in shear viscosity between adjacent layers, the shear rate along the flow path, the length of the flow path. Refer to Figure 3 for details. In this example, the green polymer is less viscous than the yellow colored polymer. The polymers change position to minimize resistance and pressure drop. This phenomenon is often referred to as **Viscous Encapsulation**.

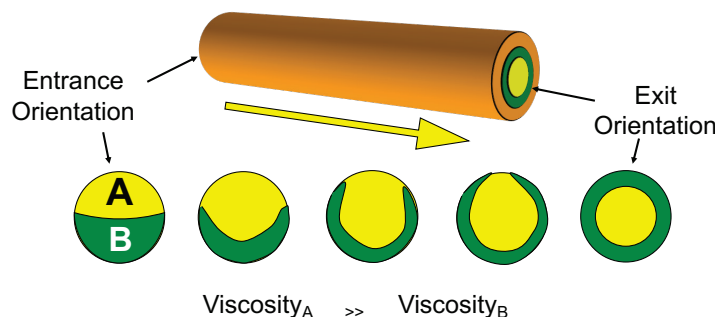


Figure 3: Polymers change relative position along melt flow path due to Viscous Encapsulation

Viscous encapsulation can occur inside single manifold cast film dies even if reasonably well distributed polymer is delivered to the die inlet. Figure 4 illustrates how a lower viscosity layer can encapsulate a higher viscosity layer, even when the distribution entering the die was balanced correctly.

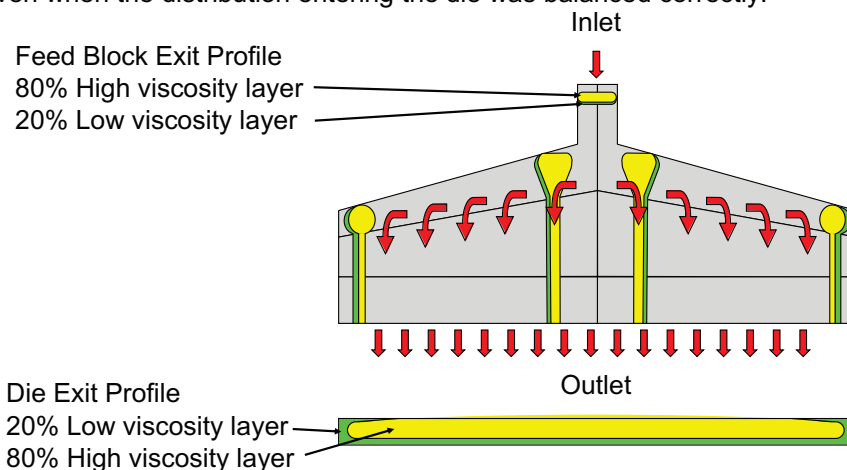


Figure 4: Polymers change relative position inside single manifold cast film

### Strategies to Prevent Very Long Wave Interfacial Instability

- |                       |  |
|-----------------------|--|
| Raw Material          | <ul style="list-style-type: none"> <li>• Modify formulation to minimize shear viscosity differences of adjacent layers.</li> <li>• Reduce COF of metal surfaces by adding polymer processing aid (PPA).</li> </ul>   |
| Processing Conditions | <ul style="list-style-type: none"> <li>• Reduce melt temperature of lower viscosity layer by adjusting extruder temperature profile. Do not exceed critical back pressure limits.</li> <li>• Increase melt temperature of higher viscosity layer by adjusting extruder temperature profile. Do not overheat to minimize thermal degradation.</li> <li>• Decrease output rate gradually to decrease shear rate in melt flow channels of adaptor and die (slower screw rotation speed). Decrease line speed to maintain target overall gauge. Ensure output rate is sufficient to avoid stagnation and thermal degradation in melt flow channels.</li> </ul> |
| Equipment             | <ul style="list-style-type: none"> <li>• For cast film dies:             <ul style="list-style-type: none"> <li>• Use adjustable die block upstream of die to compensate for viscous encapsulation. Fine tune when changing output rates or layer ratios;</li> <li>• Modify melt flow channels to reduce path length upstream from die lips (manifold area).</li> </ul> </li> </ul>  |

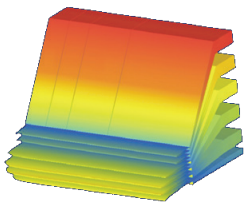
### Cast Film Die Blocks Designed to Compensate for Viscous Encapsulation

Cast film die blocks are designed to:

- Arrange the polymer streams in the desired order;
- Reshape the polymer streams for combining;
- Join the polymer streams together prior to entering the die manifold.

The 3 most common commercial feed block styles are: Layer multiplier (Dow patent); replaceable inserts; and the adjustable vane/distributor pin (Cloeren). Refer to Figures 5 and 6 for illustrations of the layer multiplier and vane/distribution pin style feed blocks.

### Layer Multiplier Feed Block



Advantages:

- Never more than 3.5% difference from an optimum flow condition.
- Flow rate balancing is dependent only on proper sequencing pin selection e.g.: direct flow to 1 channel for a 7.5% skin layer, 2 adjacent channels for a 15% skin layer
- Changing flow patterns with pin makes more consistent and repeatable product changes than changing temperatures or extruder output rates alone.

Figure 6: 3D analysis of 14 layer Dow style converging feed block

### Vane/Distributor Pin Feed Block

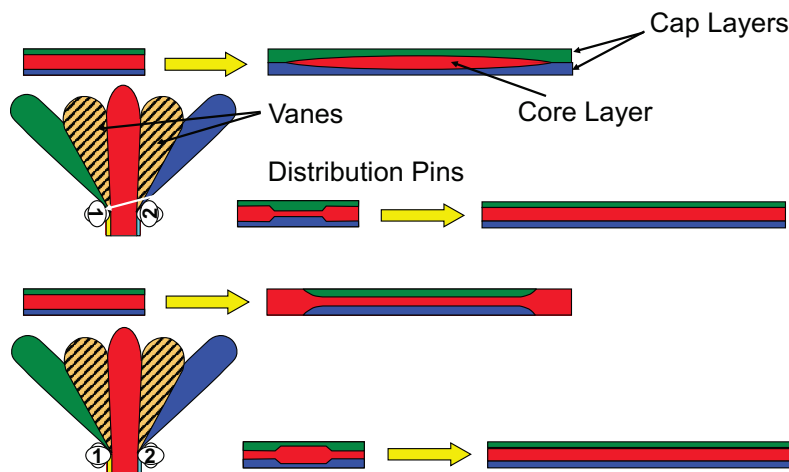


Illustration shows how unbalanced flow at die exit can be corrected. Vanes are adjusted to fine tune overall layer thickness. Distributor pins are rotated to compensate for viscous encapsulation. Fine tuning is done when line is operating at target output rate.

Figure 6. Vane/Distributor Pin die block adjustments (Cloeren)

### Long Wave Pattern Interfacial Instability

Long Wave interfacial instability occurs when one of the layers, usually the minor one, exhibits strain hardening. It can be predicted by measuring extensional viscosity of each layer. Refer to Figure 7 for a photograph on Long Wave interfacial instability and Figure 8 for an illustration of the root cause. Long Wave interfacial instability is strongly affected by the layer thickness ratio, the change in channel depth for each layer, the angle at which the layers merge and the formulation. Long wave interfacial instability frequently occurs when the minor layer is too thin. Refer to Figure 5 for an example of long wave interfacial instability patterns and Figure 6 for an illustration of its root causes.

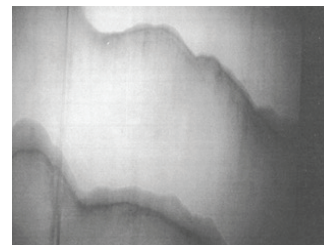


Figure 7: Long Wave interfacial instability pattern

Photograph courtesy of Compuplast North America

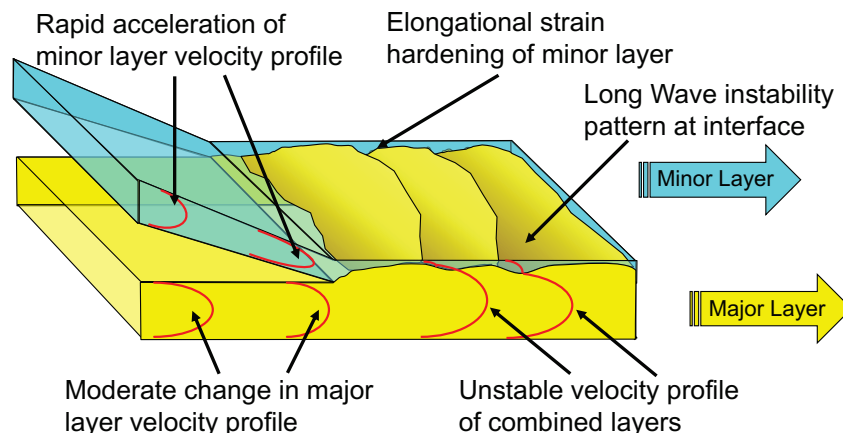


Figure 8: Root cause of Long Wave interfacial instability pattern

### Strategies to Prevent Long Wave Interfacial Instability

#### Raw Material

- Modify formulation to delay onset of strain hardening, usually of the minor layer (less highly branched molecules such as LDPE, smaller average molecular size as measured by higher melt flow rate (MFR)).
- Change the layer ratio so that the minor layer is a larger percentage of the total film structure.
- Reduce shear stress at the merge point by adding polymer processing aid (PPA) to the minor layer.

#### Processing Conditions

- Reduce extensional viscosity and stress at layer interface by increasing melt temperature of minor layer. Change in 5°C (10°F) increments and wait 15 to 20 minutes for process to stabilize. Avoid thermal degradation due to overheating.
- Decrease output rate gradually to decrease acceleration rate of the minor layer as it merges with the major layer (slower screw rotation speed). Decrease line speed to maintain target overall gauge. Avoid reducing output to rate that causes stagnation and thermal degradation of minor layer.

#### Equipment

- Decrease the channel depth of the minor layer to pre-accelerate the minor layer before it reaches the merge point.
- Decrease the merge angle at which the layers combine to minimize stress and flow direction changes at the merge point.
- Polish and replate internal die surfaces to reduce COF and shear stress at merge point.

### Zig Zag Pattern Interfacial Instability

Zig zag interfacial instability occurs when shear stress at the interface between adjacent layers exceeds critical values. It can be predicted by measuring the shear viscosity of adjacent layers. Refer to Figure 9 for an example and Figure 10 for an illustration of the root cause. The watch is included to show the scale of the pattern.

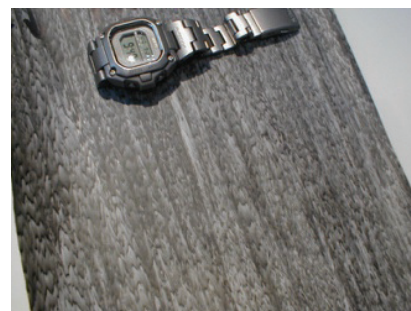


Figure 9: Zig Zag Interfacial instability pattern

Photograph courtesy of Compuplast North America

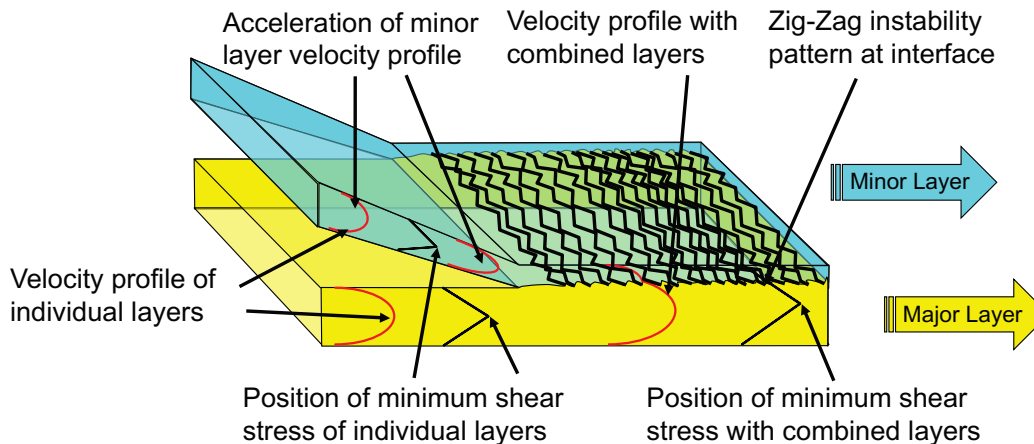


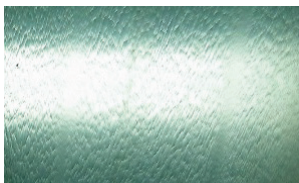
Figure 10: Root cause of Zig Zag interfacial instability pattern

### Strategies to Prevent Zig Zag Interfacial Instability

- |                       |   |
|-----------------------|---|
| Raw Material          | <ul style="list-style-type: none"> <li>• Reduce extensional viscosity and stress at layer interface by increasing melt.</li> <li>• Modify formulation to minimize shear viscosity differences of adjacent layers.</li> <li>• Change layer ratio to shift layer interface towards the center of the merged flow channel where shear stress is minimized.</li> <li>• Reduce shear stress at the merge point by adding polymer processing aid (PPA) to the minor layer.</li> </ul> |
| Processing Conditions | <ul style="list-style-type: none"> <li>• Reduce shear viscosity and stress at layer interface by increasing melt temperature of adjacent layers. Change in 10°F (5°C) increments and wait 15 to 20 minutes for process to stabilize.</li> <li>• Decrease output rate gradually to decrease shear stress at layer interface (slower screw rotation speed). Decrease line speed to maintain correct gauge.</li> </ul>   |
| Equipment             | <ul style="list-style-type: none"> <li>• Reduce coefficient of friction of die block surfaces (replace worn out plating).</li> <li>• Inspect and replace defective thermocouples or heaters in die block or die (if required).</li> </ul>   |

### Melt Fracture

Melt fracture is often referred to as ***Shark Skin*** or ***Orange Peel***. A photograph of a typical melt fracture pattern is shown in Figure 11.



- Pattern can be zig zag or random
- Often confused with interfacial instability in transparent coextruded films

Figure 11: Melt fracture surface pattern

Two mechanisms cause melt fracture. The first is slip/stick flow cycling along the melted polymer slides along metal surfaces inside the die. The second is skin rupture, which is caused by extension rates after the melted polymer leaves the die lip. Both concepts are illustrated Figure 12. Die swell occurs because the pressure inside the die is much greater than atmospheric pressure.



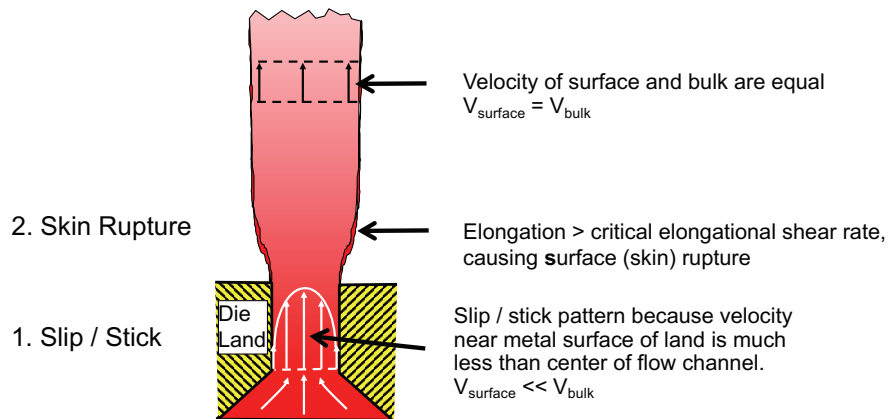


Figure 12: Melt fracture from slip / stick and skin rupture

Each polymer has its own critical shear rate above which acceptable film is not possible. Melt fracture occurs at the die lips when extruder outputs exceed the critical shear rate of the material(s). Critical shear rate and not extruder capacity is generally the bottleneck affecting the output of most polymers.

### Strategies to Prevent Melt Fracture

The principle strategy to prevent melt fracture is to reduce the shear stress at the die lip surface below the critical level. Keep in mind that wider die gaps may lead to draw resonance. Refer to Section 1.7.4 for details. When changing die lip temperatures, allow 5 to 10 minutes for the process to stabilize. When changing extruder temperature profiles, allow 15 to 20 minutes for the process to stabilize.

- |                       |   |
|-----------------------|---|
| Raw Material          | <ul style="list-style-type: none"> <li>• Reduce viscosity by selecting higher MFI blend.</li> <li>• Add polymer processing aid to reduce COF of internal die surfaces.</li> </ul>   |
| Processing Conditions | <ul style="list-style-type: none"> <li>• Increase die lip temperature in 10°F (5°C) steps.</li> <li>• Increase die body temperature in 10°F (5°C) steps.</li> <li>• Increase melt temperature by modifying extruder temperature profile.</li> <li>• Decrease output rate gradually to decrease shear stress at die lip surface (slower screw rotation speed). Decrease line speed to maintain correct gauge.</li> </ul> |
| Equipment             | <ul style="list-style-type: none"> <li>• Inspect and repair die lip heaters and thermocouples (if required).</li> <li>• Increase die gap of cast film dies by turning die bolt ¼ turn at a time. Increase line speed to maintain correct gauge.</li> <li>• Replace die pin with larger die gap (blown film dies).</li> <li>• Reduce coefficient of friction of die lip surfaces (replace worn out plating).</li> </ul>  |

A common misconception is that slip additives are as effective as processing aid but are less expensive. Most additive masterbatches use high melt index carrier resins so that the additives melt quickly and coat the other pellets. This helps improve mixing. It reduces melt fracture because adding slip masterbatch is the same as blending with a lower viscosity polymer. Problems with excessive slip concentration on the surface may result if too much is added to the formulation.

Raising the temperature at the die lips is another alternative. This can be done by raising the melt temperature, reducing viscosity, and avoiding the critical shear stress that causes melt fracture. Raising the melt temperature often reduces the maximum output capacity because most blown film lines are limited by bubble cooling capacity. Another alternative is to use heated die lips. Melt fracture may be produced in the spirals as well as at the die lips. Die lip heaters will not affect melt fracture caused inside the spirals. Die lip heaters are expensive and difficult to maintain, so they are only when absolutely necessary.

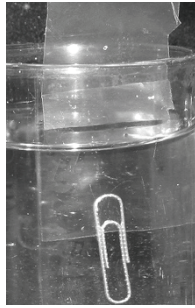
### Die Plating

Dies lips are usually plated with chrome and spirals with nickel reduce the COF and minimize adhesion of degraded polymer. Two tests to determine if the plating has worn off are commonly used. Rub the metal surface with a 400 or higher emery cloth to remove carbon residue. A grey residue indicates that steel is

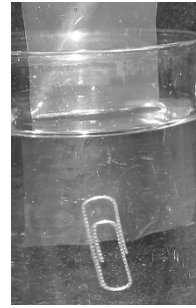
exposed (no plating remains). A silver color residue indicates that chrome is present and pale yellow indicates that nickel is present on the metal surface. An alternative is to wipe the cleaned surface with a dilute solution of copper sulphate and water. If the solution reacts with the exposed steel and changes to a pale bronze color, it indicates that no plating remains in that area.

#### **Test to Distinguish Zig Zag Interfacial Instability from Melt Fracture**

Melt fracture is only present on the outside surface of the film. Although it looks like interfacial instability in transparent films, it disappears when the film is immersed in water. Interfacial instability is like melt fracture between two layers of film. The interface between two layers does not come into contact with the water, so it remains hazy. Refer to Figure 13 for details.



Melt fracture becomes transparent



Interfacial instability remains hazy

Figure 13: Test to distinguish melt fracture from interfacial instability

#### **Conclusions**

It is critical to identify the pattern of distortion in coextruded films before selecting a strategy to eliminate the problem. In almost all cases, options will include changes to formulations, processing conditions and equipment modifications. Selecting the preferred strategy will depend on a balance between scrap rates, down time and raw material costs.